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Single shot time stamping of ultrabright radio frequency compressed electron pulses

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We demonstrate a method of time-stamping Radio Frequency compressed electron bunches for Ultrafast Electron Diffraction experiments in the sub-pC regime. We use an *in-situ* ultra-stable photo-triggered streak camera to directly track the time of arrival of each electron pulse and correct for the timing jitter in the radio frequency synchronization. We show that we can correct for timing jitter down to 30 fs root-mean-square with minimal distortion to the diffraction patterns, and performed a proof-of-principle experiment by measuring the ultrafast electron-phonon coupling dynamics of silicon. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4813313]

Pulsed electron and X-ray sources have recently matured to a state where many ultrafast structural phenomena in crystalline matter can be monitored with sufficient spatial and temporal resolution to fully resolve the key modes in transitions to extreme states of matter,[1–3] structure order parameters in strongly correlated electron lattices,[4–6] and molecular motions in organic crystals.[7] In particular, X-FELs (X-ray Free Electron Lasers), radio frequency (RF) accelerated,[8–10] and RF compressed[11–13] electron sources have recently emerged as the flagship technologies in the field, boosting the probe brightness by orders of magnitude to enable entire crystal projections to be captured in a single pulse. However, RF based sources still suffer from a major complication: the synchronization between the laser oscillator and RF electronics. State of the art free electron laser facilities such as the Linac Coherent Light Source (LCLS) still report nominal timing jitter between laser and X-ray of 100–200 fs RMS,[14] and RF compressed electron sources have reported similar quantities.[12,13] However, work at LCLS has recently demonstrated that timing jitter can be mitigated by several single shot time stamping techniques.[14–18] With electrons, electro-optic sampling techniques can be used in a similar fashion for bunch charges more than a few pC.[19] For the typical fC bunches used in UED (Ultrafast Electron Diffraction), no current technique is sensitive enough to determine the arrival time of individual pulses with respect to the excitation laser. Using a high-sensitivity, ultrastable photo-triggered streak camera,[20] we report time stamping of 50 fC RF compressed electron bunches in diffraction mode with 30 fs RMS resolution, and demonstrate its ability to significantly improve the Instrument Response Function (IRF) and fidelity in a time-resolved electron diffraction experiment of single crystalline silicon.

Figure 1 depicts the experimental setup. The electron pulses are generated via photo-emission from the surface of a 20 nm thin gold photo-cathode using 270 nm UV pulses derived by frequency tripling from a home built Ti-Sapphire laser. A 50 fC electron bunch is then accelerated at 95 keV through a 1 cm gap towards a silicon anode with a 700 μm aperture. The bunch is then collimated by a magnetic lens (M1) into the RF rebunching cavity. A sinusoidally time varying electric field synchronized with the arrival of the electron pulse inverts the velocity chirp of the bunch as it passes through the cavity, allowing it to come to a temporal focus downstream at the sample position.[21] A second magnetic lens (M2) is used to compensate for the spatial defocusing effects of the RF cavity. Less than 1 mm after the sample, the electron pulse enters a compact, high streak velocity, photo-triggered streak camera:[20] a pair of parallel plates (3 mm × 3 mm) with 200 μm gap charged with a 60 ns, 1200 V high voltage pulse. A trigger pulse precisely synchronized with the pump pulse by deriving it from the original pulse via a beam-splitter impinges on the photo-switch and discharges the streak plates, setting off a 5.5 GHz inductance-resistance-capacitance (LRC) circuit oscillation.

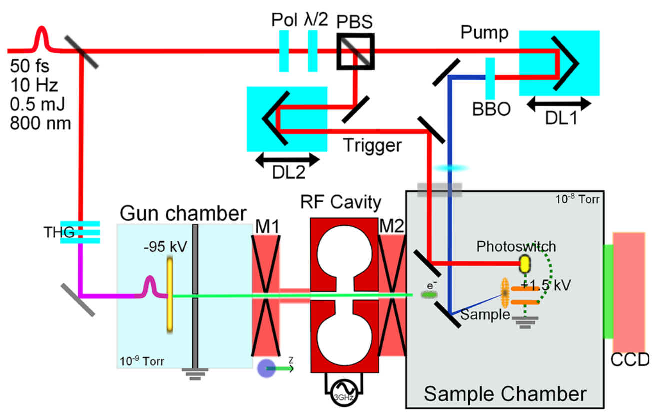

FIG. 1. Detailed schematic of the experimental setup including streak camera. Photoelectrons are produced by back-illumination of a gold cathode (z = 0 cm). They are then accelerated at 95 keV towards a 700 μm aperture in a disk silicon anode wafer (z = 1 cm). A magnetic lens (M1, z = 9 cm) collimates the bunch before entering the RF rebunching cavity (z = 39 cm), and a second magnetic lens (M2, z = 45 cm). The electrons then come to a temporal focus at the sample (z = 59 cm) and diffract from it. The electrons then pass through a pair of high voltage streak plates, placed 1 mm from the sample, encoding their time of arrival information with respect to a trigger laser pulse. They are then recorded at the CCD detector (z = 85 cm).

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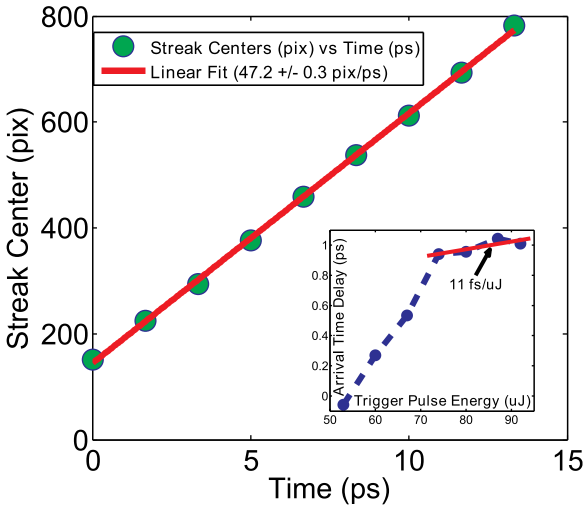

FIG. 2. Characterization of the streak camera. The streak velocity is determined by changing the trigger delay and measuring the electron displacement on the CCD, yielding a value of 47 pixels/ps (2.8 mrads/ps) with error ($\pm$0.3 pixel per/ps). Inset: The streak velocity and arrival time is also affected by trigger pulse energy. The energy is given for 800 nm pulses with beam diameter of 3 mm FWHM. We worked in a saturated regime at 80 μJ where a change in trigger pulse energy makes a minute contribution to the arrival time (11 fs/μJ).

The electrons then experience a transverse electric field whose magnitude is directly proportional to the arrival time with respect to the trigger pulse. The diffraction pattern is then recorded on the CCD, with the time of arrival information encoded automatically in the transverse displacement of the beam.

The streak camera has been used previously to measure sub-150 fs FWHM temporal duration of short electron pulses[20] and demonstrated sub-ps lattice dynamics in streaking mode.[22] Its compact design, high frequency of operation, and laser triggering mechanism allow for extremely low shot-to-shot timing jitter, previously estimated to be less than 50 fs FWHM.[20,22] Previous experiments have used the streak camera in accumulation mode, but the high brightness of RF compressed electron pulses naturally allow the streak displacement to be measured in a single shot. These two properties together make it the ideal tool for single shot time stamping of RF compressed electron bunches, typically highly sensitive to laser-RF synchronization jitter.[12,13] As shown in Fig. 1, we installed the streak camera <1 mm after the sample so that the entire diffraction pattern is effectively streaked onto the CCD. Due to the temporal bunching of the electrons, very little streaking actually occurs, leaving the diffraction pattern mostly unperturbed. However, the entire pattern is shifted parallel to the streaking direction, the magnitude of which depends on the arrival time of the electron pulse with respect to the streak trigger. The center of the 0th order spot is then used to "time-stamp" the relative arrival time of each diffraction pattern.

First, we characterized the streak velocity in the setup and minimized the intrinsic jitter due to intensity noise in the trigger pulse. We measured the streak velocity by varying the delay of the trigger arm and recording the displacement of the electrons on the CCD. We reported a streak velocity of 47 pixels/ps on the CCD corresponding to 2.8 mrads/ps shown in Fig. 2. The resistance of the photoswitch depends on the trigger pulse energy and can change the phase the LRC oscillation and the voltage that the electrons see as a result.[20] Therefore, we verified that we worked in a saturated regime to minimize this effect. The inset to Fig. 2 shows that with a trigger pulse energy above 50 μJ, the streak camera becomes highly insensitive to changes in trigger energy leading to a slope of only 11 fs/μJ. We choose to use a pulse energy of 80 μJ (with beam diameter of 3 mm FWHM), giving a sensitivity of 9 fs/1% change in laser intensity. We measured the intensity fluctuations of the output of our laser with a photodiode, yielding <1 %/hr RMS and <0.3 %/s RMS. Thus, with slow active feedback, the noise on the arrival time caused by intensity fluctuations could be negligible.

We then used the streak camera to determine the timing characteristics of our RF gun. The temporal IRF of the system was previously measured by ponderomotive scattering and reported to be ≈300–500 fs FWHM in two independent measurements.[12,13] Lack of single shot stability in the ponderomotive signal strongly suggested that timing jitter between the RF and the laser is a significant contribution of the measured temporal IRF. We first confirmed this with the streak camera by measuring the streak displacement of a

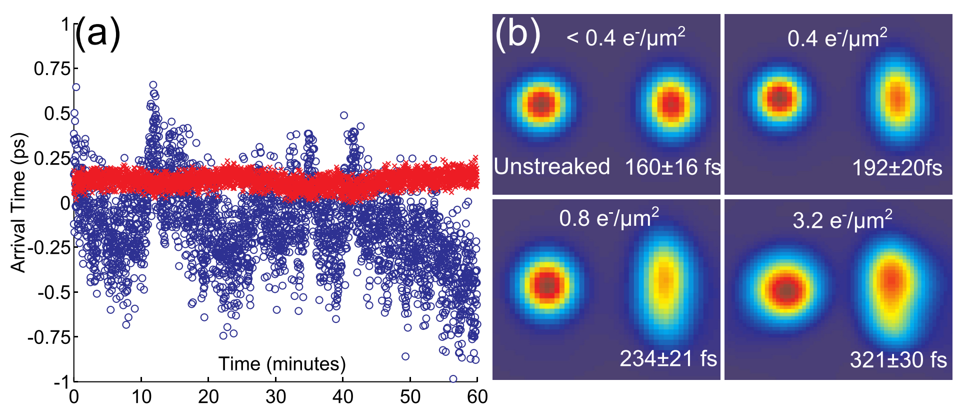

FIG. 3. Characterization of the RF compressed electron pulses using the streak camera. (a) Plot shows single shot electron arrival times measured by the streak camera. Using the experimentally determined streak velocity in Fig. 2, the jitter (blue trace) is 200 fs RMS. The reference direction (red trace), experiencing no streak voltage, yields a minimal temporal resolution to the determination of the arrival time of 30 fs RMS. (b) The panels show the minimal pulse duration achievable for the given electron densities at the sample position. Each pair of beams represents an unstreaked (left) and streaked (right) pulse on the CCD for a given measured electron density at the sample position. Pulse durations are given in FWHM. The scaling for each image pair is different.

series of single shot bunches (Fig. 3(a)). We determined the streak displacements by computing the Gaussian fit of the center of each pulse. In the streaked direction (blue trace), the jitter of the streak center is 10 pixels RMS, which implies the arrival time jitter is 200 fs RMS (based on 47 pixel/ps streak velocity), closely agreeing with our previous results using ponderomotive scattering.[12] As a reference, the displacement centers in the unstreaked direction (red trace) is also measured and found to have a RMS of 1.5 pixels or 30 fs RMS. Measuring the displacement in the streaking direction but with voltage off yields identical results. This figure imposes an upper bound on the resolution of the arrival time, limited by both the center determination error of the fits and the electron beam pointing instabilities in the gun. Note that the streak center determination can be much more accurate than the typical streak impulse response, which is limited by the transverse width of beam on the CCD.

We then characterized the pulse length of the RF compressed bunches as a function of electron density at the sample position. Fig. 3(b) shows the minimal temporal durations of single electron bunches derived from measuring the streak lengths through a 50 μm aperture and then calculating the deconvolution with an aperture function.[20] This is done by tuning the lens after the RF cavity (M2 in Fig. 1 controls the electron density at the sample) and then tuning the RF amplitude to find the shortest bunch. The minimal pulse width increased with strong focusing conditions, indicative that transverse space charge effects still play a significant role in the current RF rebunching scheme. At lower densities, pulse durations between 100 and 200 fs FWHM can be produced. However, at electron density of 3.2 $e^-/\mu\text{m}^2$ at the sample position, the pulse duration is limited to $\approx$300 fs FWHM. The electron density is calculated as $N_e/(\pi * (\text{FWHM}/2)^2)$ where $N_e = 330\,000$ and the FWHM beam width is measured with a knife edge at the sample position without the aperture.

To demonstrate the feasibility of the time-stamping technique, we measured the well-studied fast lattice-heating dynamics of Si, using a 30 nm (001) oriented sample.[23,24] Fig. 4(a) shows the silicon diffraction pattern passing through the streak plates with no streak voltage and Fig. 4(b) shows the same pattern but with streak voltage (streak velocity of 47 pixels/ps). The pattern is clipped near the edges due the current widths of the streak plates. A 400 nm, 50 fs FWHM, 2.2 mJ/cm^2 laser pulse is used to excite the single crystal sample. Single shot electron pulses at 2 Hz with 330 000 electrons per bunch and 360 μm FWHM spot size (no aperture) (3.2 $e^-/\mu\text{m}^2$) were used to probe the structural changes. To increase the fidelity of the measurements, we used TDI (Time Delayed Integration) mode to capture multiple single-shot diffraction patterns on one CCD Image, in a timed sequence. Fig. 5(a) shows 5 separate identical traces that were generated with 3 h separation (total of 15 h), showing clear arrival time instability due to jitter and longer term temperature drift still present in the system. In each trace, every time point represents the average of 150 single electron shots. Now using the identical data set in Fig. 5(a), and only the center positions of the 0th order peaks, we can re-bin all the electron shots according to their actual arrival time based on the streak velocity and their distances from the origin, resulting in Fig. 5(b). No other method of data manipulation was used. The improvement in signal to noise and resolution of the dynamics is clearly demonstrated in this comparison.

The advantage of time stamping is two-fold:

1. In a typical UED experiment using RF compression, the laser-electron t_0 must be measured frequently to make sure it hasn't drifted far (i.e., once every hour). Therefore, scans must be kept short and recombined

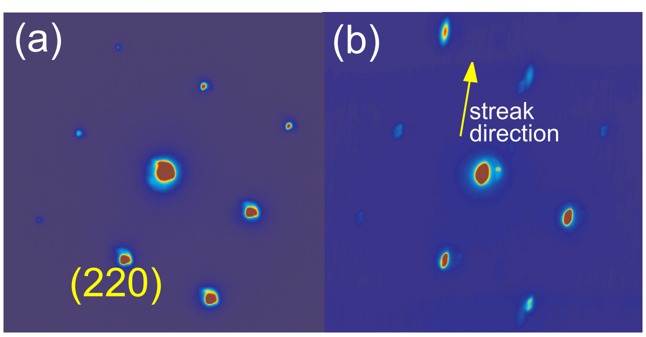

FIG. 4. Comparison of the streaked and unstreaked diffraction patterns. (a) Panel shows an averaged diffraction pattern of a Si(001) oriented thin crystal and panel (b) shows the same sample but with the streak camera turned on.

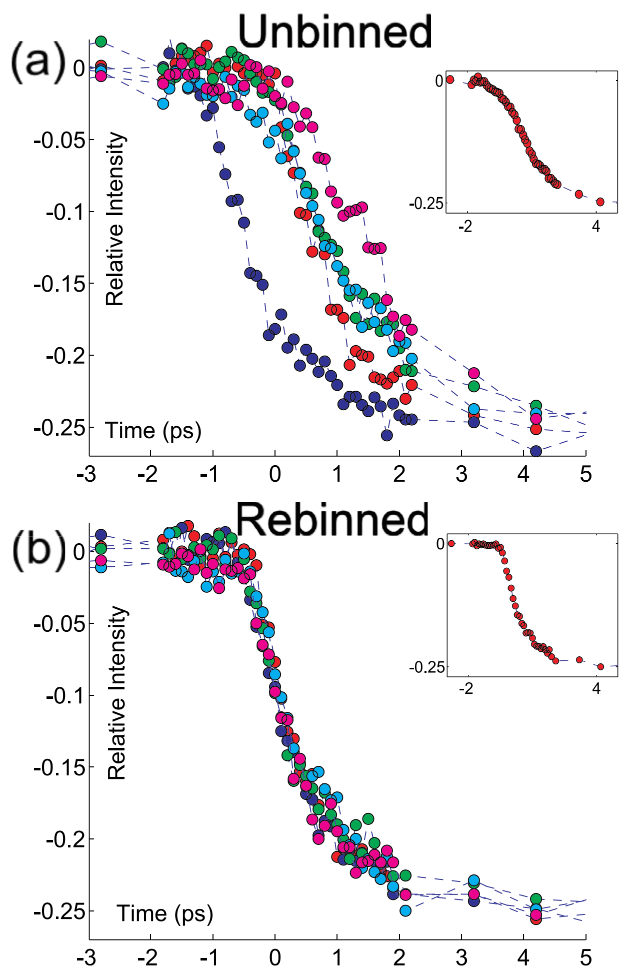

FIG. 5. Proof of principle experiment with time stamping. (a) The (001) oriented silicon sample is pumped with 400 nm light. The photoinduced relative change to the (220) bragg peak is plotted as a function of time. Five traces were taken at 3 h time intervals, showing the effects of phase drift masking the dynamics. (b) The same data but with each shot re-binned using the time stamp generated from the displacement of the streaked diffraction patterns. The insets show the average of the 5 traces in (a) and (b), respectively.

later and "bad" scans must be manually rejected. This makes data collection very inefficient, especially small signals that do not readily enable obvious t_0 determination. By time stamping every pulse, the fidelity of the data stays consistent indefinitely (i.e., 15 h in Fig. 5(b)) and even the smallest signal can be recovered this way.

2. The temporal resolution can be increased by negating the effects of jitter on the IRF. Although the decay of the Si bragg peak is too slow to test the true limits of the temporal resolution, the high level of agreement between the 5 traces in Fig. 5(b) illustrates the very low temporal jitter.

In this letter we have presented the first demonstration of time stamping of sub-pC RF compressed electron bunches with clearly demonstrated increased temporal fidelity and reduction of the timing jitter down to 30 fs RMS in UED experiments. A streak camera with higher streak voltage and larger gap spacing is being developed to allow for more diffraction orders to be captured. We are also using the streak camera to aid in the optimization of the RF compression system to further increase the spatial-temporal density of the electron bunches towards an overall sub-100 fs IRF. In addition, we are investigating the prospect of extending the application of the photo-triggered streak camera to relativistic electron sources.

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